

[Note: with the publication of the Fifth Edition of AP-42, the Chapter and Section number for Sulfur Recovery changed to 8.13.]

BACKGROUND REPORT

AP-42 SECTION 5.18

SULFUR RECOVERY

Prepared for

**U.S. Environmental Protection Agency
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AP-42 Background Report

TECHNICAL SUPPORT DIVISION

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Air Quality Planning and Standards
Research Triangle Park, NC 27711

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1.0 INTRODUCTION

The document "Compilation of Air Pollutant Emission Factors" (AP-42) has been published by the U.S. Environmental Protection Agency (EPA) since 1972. Supplements to AP-42 have been routinely published to add new emission source categories and to update existing emission factors. AP-42 is routinely updated by the EPA to respond to new emission factor needs of the EPA, state and local air pollution control programs, and industry.

An emission factor relates the quantity (weight) of pollutants emitted to a unit of activity of the source. The uses for the emission factors reported in AP-42 include:

1. Estimates of area-wide emissions;
2. Emission estimates for a specific facility; and
3. Evaluation of emissions relative to ambient air quality.

The purpose of this report is to provide background information obtained from industry comment and 8 test reports to support revision of the process description and/or emission factors for sulfur recovery.

Including the introduction (Chapter 1), this report contains four chapters. Chapter 2 gives a description of the sulfur recovery industry. It includes a characterization of the industry, an overview of the different process types, a description of emissions, and a description of the technology used to control emissions resulting from sulfur recovery.

Chapter 3 is a review of emissions data collection and analysis procedures. It describes the literature search, the screening of emission data reports, and the quality rating system for both emission data and emission factors. Chapter 4 details criteria and noncriteria pollutant emission factor development. It includes the review of specific data sets and the results of data analysis. Appendix A presents AP-42 Section 5.18.

2.0 INDUSTRY DESCRIPTION

2.1 GENERAL

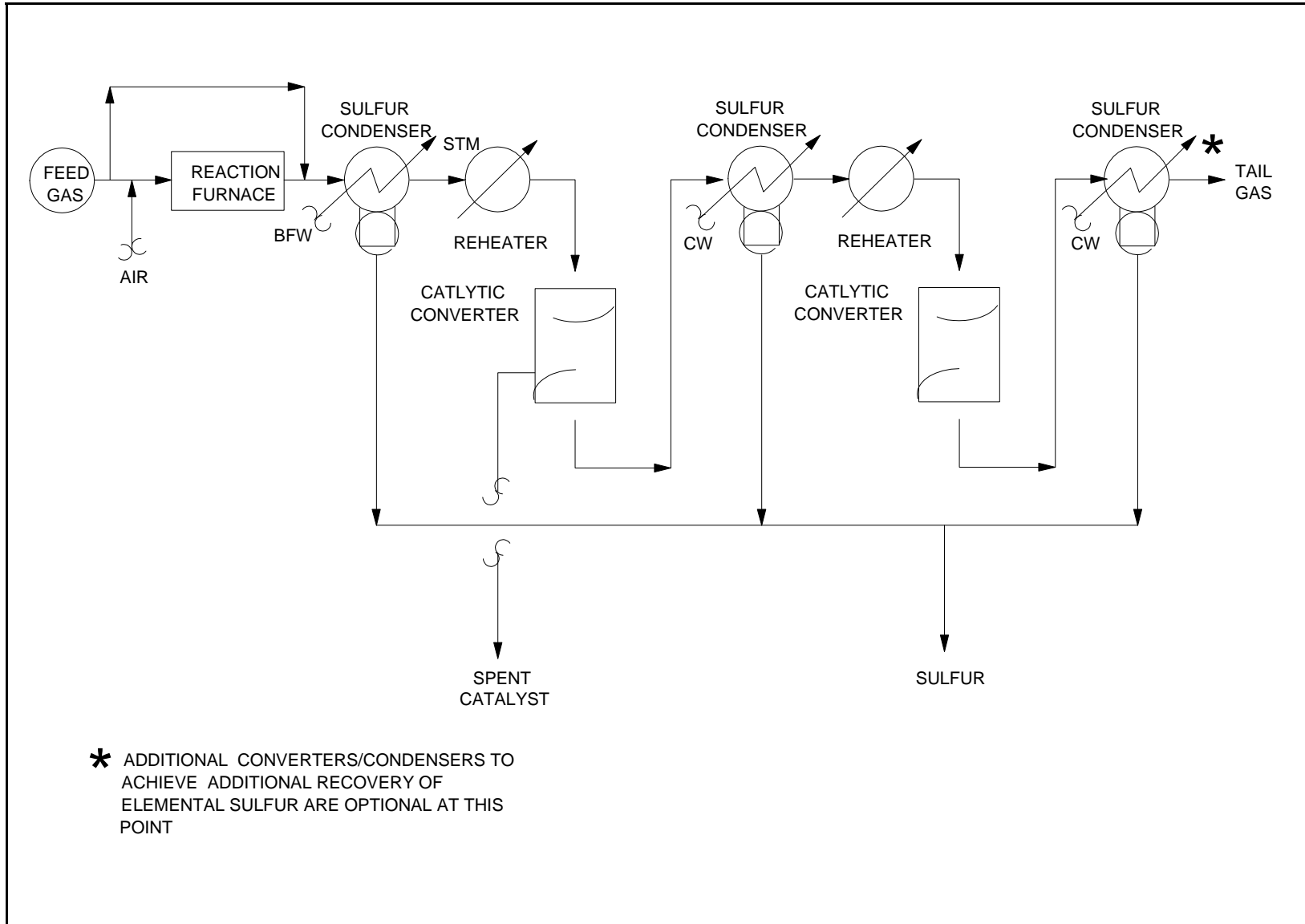
Sulfur recovery refers to the conversion of hydrogen sulfide (H_2S) to elemental sulfur. Hydrogen sulfide is a byproduct of processing natural gas and refining high-sulfur crude oils. The most common conversion method used is the Claus process. Approximately 90 to 95 percent of recovered sulfur is produced by the Claus process. The Claus process typically recovers 95 to 97 percent of the hydrogen sulfide feedstream.

Over 5.9 million megagrams (6.5 million tons) of sulfur were recovered in 1989, representing about 63 percent of the total elemental sulfur market in the U.S. The remainder was mined or imported. The average production rate of a sulfur recovery plant in the U.S. varies from 51 to 203 megagrams (56 to 224 tons) per day. There are over 250 Claus sulfur plants operating in petroleum refineries and onshore natural gas processing facilities.

2.2 PROCESS DESCRIPTION

Hydrogen sulfide occurs naturally in crude oil and natural gas. It is removed and converted to elemental sulfur by the Claus process. Figure 2.2-1 shows a typical Claus sulfur recovery unit.

Figure 2.2-1 Typical Claus sulfur recovery unit



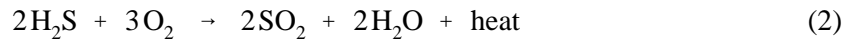
The process consists of multistage catalytic oxidation of hydrogen sulfide according to the following overall reaction:



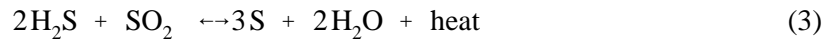
Each catalytic stage consists of a gas reheater, a catalyst chamber, and a condenser.

There are four main variations of the Claus process which differ primarily in the way in which the heat balance is maintained. These are the straight through process, the split flow process, the split flow process with preheating of feed stream, and the sulfur recycle process. The straight through process is utilized when the H_2S concentration in the acid gas feed is greater than 50 mole percent. The split flow process configuration is used when the H_2S concentration in the feed is between 20 and 50 mole percent. If the feed stream contains less than 20 mole percent H_2S and the flame is not self-sustaining at ambient temperature, the split flow scheme with preheat is employed. The sulfur recycle process is used when the feed contains less than 10 volume percent H_2S and a self-sustaining flame occurs.

The Claus process involves burning one third of the hydrogen sulfide (H₂S) with air in a reactor furnace to form sulfur dioxide (SO₂) according to the following reaction:



The remaining uncombusted two thirds of the hydrogen sulfide undergoes Claus reaction (reacts with SO₂) to form elemental sulfur as follows:



The furnace normally operates at temperatures ranging from 980 to 1540°C (1800 to 2800°F) with pressures rarely higher than 70 kPa (10 psia). Hot gas from the furnace is quenched in a waste heat boiler that generates high to medium pressure steam. About 80 percent of the heat released from burning H₂S is recovered as useful energy. Liquid sulfur from the condenser runs through a seal leg into a covered pit from which it is pumped to trucks or railcars for shipment to end users. Approximately 65 to 70 percent of the sulfur is recovered. The cooled gases exiting the condenser are then sent to the catalyst beds.

The catalytic reactors operate at lower temperatures than the furnace, ranging from 200 to 315°C (400 to 600°F). Alumina or bauxite is sometimes used as a catalyst. Because this reaction represents an equilibrium chemical reaction, it is not possible for a Claus plant to convert 100 percent the incoming sulfur compounds to elemental sulfur. Therefore, two or more stages are used in series to recover the sulfur. Each catalytic stage can recover one-half to two-thirds of the incoming sulfur. The number of catalytic stages depends upon the level of conversion required. It is estimated that 95 to 97 percent overall recovery can be achieved, depending on the number of catalytic reaction stages and the type of reheating method used. If the sulfur recovery unit is located in a natural gas processing plant, the type of reheat employed is typically either auxiliary burners or heat exchangers, with steam reheat being used occasionally. If the sulfur recovery unit is located in a crude oil refinery, the typical reheat scheme uses 3536 to 4223 kPa (500 to 600 psig) steam for reheating purposes. Most plants are now built with two catalytic stages, although some air quality jurisdictions require three. From the condenser of the final catalytic stage, the process stream passes to some form of tailgas treatment process. The tailgas, containing H₂S, SO₂, sulfur vapor, and traces of other sulfur compounds formed in the combustion section,

escapes with the inert gases from the tail end of the plant. Thus, it is frequently necessary to follow the Claus unit with a tailgas cleanup unit to achieve higher recovery.

In addition to the oxidation of H_2S to SO_2 and the reaction of SO_2 with H_2S in the reaction furnace, many other side reactions occur in the furnace. Three of these side reactions are:



2.3 EMISSIONS AND CONTROLS

Emissions from the Claus process are directly related to the recovery efficiency. Higher recovery efficiencies mean less sulfur compounds are emitted in the tailgas. Older plants, and very small Claus plants producing less than 20 megagrams (22 tons) per day of sulfur without tailgas cleanup, have varying sulfur recovery efficiencies. The efficiency depends upon several factors, including the number of catalytic stages, the concentrations of H_2S and contaminants in the feed stream, stoichiometric balance of gaseous components of the inlet, operating temperature, and catalyst maintenance.

A two-bed catalytic Claus plant can achieve 94 to 96 percent efficiency. Recoveries range from 96 to 97.5 percent for a three-bed catalytic plant and range from 97 to 98.5 percent for a four-bed catalytic plant. At normal operating temperatures and pressures, the Claus reaction is thermodynamically limited to 97 to 98 percent recovery. Tailgas from the Claus plant still contains 0.8 to 1.5 percent sulfur compounds. A typical Claus unit tailgas composition before application of control devices is as follows: 86.18 volume percent nitrogen, 11.57 percent CO_2 , 1.22 percent argon, 0.42 percent hydrogen, 0.22 percent hydrogen sulfide (H_2S), 0.15 percent carbonyl sulfide (COS), 0.13 percent SO_2 , and 0.11 percent carbon disulfide (CS_2).

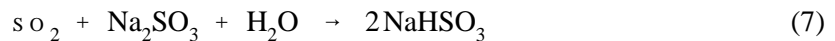
Emissions from the Claus process may be reduced by: 1) extending the Claus reaction into a lower temperature liquid phase, 2) adding a scrubbing process to the Claus exhaust stream, or 3) incinerating the hydrogen sulfide gases to form sulfur dioxide.

Currently, there are five processes available that extend the Claus reaction into a lower temperature liquid phase including the BSR/Selectox, Sulfreen, Cold Bed Absorption, Maxisulf, and IFP-1 processes. These processes take advantage of the enhanced Claus conversion at cooler

temperatures in the catalytic stages. All of these processes give overall sulfur recoveries of 98 to 99 percent when installed downstream of a typical two- or three- stage Claus sulfur recovery unit, and therefore emit lower sulfur emissions.

Sulfur emissions can also be reduced by adding a scrubber at the tail end of the plant. There are essentially two generic types of tailgas scrubbing processes: oxidation tailgas scrubbers and reduction tailgas scrubbers. The oxidation scrubbing process is used to scrub sulfur dioxide (SO₂) from incinerated tailgas and recycle the concentrated SO₂ stream back to the Claus process. There are at least three oxidation scrubbing processes: the Wellman-Lord, Stauffer Aquaclaus, and IFP-2. Only the Wellman-Lord process has been successfully applied to U.S. refineries.

The Wellman-Lord process uses a wet, generative process to reduce stack gas sulfur dioxide concentration to less than 250 parts per million volume (ppmv) and can achieve approximately 99.9 percent sulfur recovery. Claus plant tailgas is incinerated and all sulfur species are oxidized to form sulfur dioxide (SO₂) in the Wellman-Lord process. Gases are then cooled and quenched to remove excess water and to reduce gas temperature to absorber conditions. The rich SO₂ gas is then reacted with a solution of sodium sulfite (Na₂SO₃) and sodium bisulfite (NaHSO₃) to form the bisulfite:



The offgas is reheated and vented to the atmosphere. The resulting bisulfite solution is boiled in an evaporator-crystallizer, where it decomposes to SO₂ and H₂O vapor and sodium sulfite is precipitated:



Sulfite crystals are separated and redissolved for reuse as lean solution in the absorber. The wet SO₂ gas is directed to a partial condenser where most of the water is condensed and reused to dissolve sulfite crystals. The enriched SO₂ stream is then recycled back to the Claus plant for conversion to elemental sulfur.

In the reduction type scrubbing process, sulfur in the tailgas is converted to H₂S by hydrogenation in a reduction step. After hydrogenation, the tailgas is cooled and water is removed. The cooled tailgas is then sent to the scrubber for H₂S removal prior to venting. There

are at least four reduction scrubbing processes developed for tailgas sulfur removal: Beavon, Beavon MDEA, SCOT and ARCO. In the Beavon process, H_2S is converted to sulfur outside the Claus unit using a lean H_2S -to-sulfur process (the Strefford process). The other three processes utilize conventional amine scrubbing and regeneration to remove H_2S and recycle back as Claus feed.

Emissions from the Claus process may also be reduced by incinerating sulfur-containing tailgases to form sulfur dioxide. In order to properly remove the sulfur, incinerators must operate at a temperature of 650°C ($1,200^\circ\text{F}$) or higher if all the H_2S is to be combusted. Proper air-to-fuel ratios are needed to eliminate pluming from the incinerator stack. The stack should be equipped with analyzers to monitor the SO_2 level.

An existing New Source Performance Standard (NSPS) limits sulfur emissions from Claus sulfur recovery plants of greater than 20.32 megagrams (22.40 ton) per day capacity to 0.025 percent (250 ppmv) by volume (Reference 3). This limitation is effective at zero percent oxygen on a dry basis if emissions are controlled by an oxidation control system or a reduction control system followed by incineration. This is comparable to the 99.8 to 99.9 percent control level for reduced sulfur.

2.4 REVIEW OF REFERENCES FOR CHAPTER 2

Pacific Environmental Services (PES) contacted the following sources to obtain the most up-to-date information on process descriptions and emissions for this industry:

- 1) Alabama Department of Environmental Management: Montgomery, AL.
- 2) Amoco Oil: Texas City, TX.
- 3) Collett Ventures Company: Chatom, AL.
- 4) Exxon Corporation: New Orleans, LA.
- 5) Florida Department of Environmental Regulation: Tallahassee, FL.
- 6) Georgia Department of Natural Resources: Atlanta, GA.
- 7) Hill Petroleum Company: Texas City, TX.
- 8) Hunt Refining Company: Tuscaloosa, AL.
- 9) Kansas Department of Health and Environment: Topeka, KS.
- 10) Michigan Department of Natural Resources: Lansing, MI.
- 11) Missouri Department of Natural Resources: Jefferson City, MO.
- 12) Mobil Exploration and Producing U.S., Inc.: Coden, AL.

- 13) Pennsylvania Department of Natural Resources: Harrisburg, PA.
- 14) Phillips Petroleum Company: Chatom, AL.
- 15) Union Oil Company: Beaumont, TX.
- 16) EPA Emission Measurement Branch (EMB), Research Triangle Park, NC.

Of the 16 sources contacted, nine (Sources #2, 3, 4, 7, 8, 12, 14, 15 and 16) responded. Exxon Corporation (Source #4) sent a summary of the test data performed at their Flomaton treating facility. The report did not have any raw field data, calibration data, or a description of the testing procedure. Therefore, it could not be used to modify the existing emission factors. Hill Petroleum Company (Source #7) sent three source test reports. However, two of the reports did not include the production rate data needed to calculate the emission factors. Union Oil Company (Source #15) also sent one test report with no production rate data. EMB (Source #16) provided a list of emission test reports related to Claus sulfur recovery retrieved from TSAR database.

PES found nine reports related to sulfur recovery in the TSAR database. Only eight of them are emission test reports conducted from 1974 to 1981. Seven of the reports did not provide production rates and therefore could not be used to revise the emission factors. PES was able to use only one of these reports (Getty Oil Company) to revise the controlled sulfur dioxide emission factor for a three-stage catalytic unit.

A total of eight source test reports from seven sources were reviewed to determine new emission factors. Hunt Refining Company (Source #8) sent two source test reports. These reports utilized different types of control devices: Shell Claus Offgas treatment (SCOT)-absorber-incinerator (Sources #2, 7 and 12) and incinerator (Sources #3, 8, 14 and 16). Only four of these reports (Collet Ventures, Mobil Exploration, Phillips Petroleum Company, and Getty Oil Company) provided information on the number of catalytic reaction stages used during the tests. Sulfur dioxide (SO₂) emission factors from a sulfur recovery plant vary depending on the number of catalytic stages used. Each catalytic reaction stage used results in different sulfur recovery efficiencies. Even processes with the same number of catalytic reaction stages may have different sulfur recovery efficiencies. PES has incorporated the information from these four reports into the AP-42 section revision by developing new emission factors. The rest of the reports are discussed only as background information.

In addition to the new source tests used to develop new emission factors, PES obtained References 1, 2, and 3 through a literature search, NTIS and by obtaining a copy of a draft AP-40 chapter on sulfur recovery. Reference 4 was obtained from the background files for the previous revision of Section 5.18 (February 1980). Reference 5 was also obtained from the background files. However, it was not listed in the references in the 1980 version.

Reference #1: "Sulfur Recovery Technology"

This article, obtained from the *Energy Progress* journal, provided a description of the general Claus process, an estimate of the efficiency of the Claus process, and a description of several extended processes used specifically at petroleum refineries. However, it does not describe any emissions or control devices used to reduce the emissions.

Reference #2: Written communication from Bruce Scott

Mr. Bruce Scott wrote a chapter on sulfur recovery for the current revision effort on AP-40. PES has obtained a copy of the chapter from the author. This chapter was used to determine the amount of sulfur recovered from the byproduct of the crude oil and natural gas processing. This reference was also used in the general process discussion and to describe the chemical reactions that occur during the sulfur recovery process.

Reference #3: Review of New Source Performance Standards for Petroleum Refinery Claus Sulfur Recovery Plants

PES obtained this reference from NTIS. It was used in conjunction with Reference 2 to describe several extended Claus processes used to reduce sulfur emissions.

Reference #4: Standards Support and Environmental Impact Statement, Volume 1: Proposed Standards of Performance for Petroleum Refinery Sulfur Recovery Plants

This document was obtained from the EPA background file for the previous revision of Section 5.18 (February 1980) and used to support the emissions and controls section.

Reference #5: "Abating Sulfur Plant Tail Gases"

This article, obtained from the *Pollution Engineering* journal, provided percentages of sulfur recovered from uncontrolled Claus plant with two, three and four catalytic stages. The percents of sulfur recovery were then used to determine sulfur dioxide emission factors.

2.5 REFERENCES FOR CHAPTER 2

1. B. Goar et al., "Sulfur Recovery Technology," *Energy Progress*, Vol. 6(2): 71-75, June, 1986.
2. Written communication from Bruce Scott, Bruce Scott, Inc. to David Hendricks, Pacific Environmental Services, Inc., February 28, 1992.
3. Review of New Source Performance Standards for Petroleum Refinery Claus Sulfur Recovery Plants, EPA-450/3-83-014, U.S. Environmental Protection Agency, Research Triangle Park, NC, August 1983.
4. Standards Support and Environmental Impact Statement, Volume 1: Proposed Standards of Performance for Petroleum Refinery Sulfur Recovery Plants. EPA-450/2-76-016a, U.S. Environmental Protection Agency, Research Triangle Park, NC, September 1976.
5. D.K. Beavon, "Abating Sulfur Plant Tail Gases," *Pollution Engineering*, January/February 1972, pp. 34-35.

3.0 GENERAL EMISSION DATA REVIEW AND ANALYSIS PROCEDURES

3.1 LITERATURE SEARCH AND SCREENING OF SOURCE TESTS

The first step of this investigation involved a search of available literature relating to criteria and noncriteria pollutant emissions associated with sulfur recovery production. This search included the following references:

- 1) AP-42 background files maintained by the Emission Factor and Methodologies Section.
- 2) References in the National Technical Information Service (NTIS).
- 3) The EPA databases, which included the *VOC/Particulate Matter (PM) Speciation Database Management System (SPECIATE)*, the *Crosswalk/Air Toxic Emission Factor Data Base Management System (XATEF)*, and the Emission Measurement Technical Information Center's *Test Methods Storage and Retrieval System (TSAR)*.

To reduce the amount of literature collected to a final group of references pertinent to this report, the following general criteria were used:

1. Emissions data must be from a primary reference; i.e., the document must constitute the original source of test data.
2. The referenced study must contain test results based on more than one test run.
3. The report must contain sufficient data to evaluate the testing procedures and source operating conditions (e.g., one-page reports were generally rejected).

Two reference documents taken from background files and NTIS references were used to revise the process description and emission controls sections in the AP-42 document. Neither document provided any emission data associated with sulfur recovery production. PES also was unable to find any emission data in the SPECIATE and XATEF. However, TSAR database listed nine reports related to sulfur recovery. Only eight of them were emission test reports conducted from 1974 to 1981. Seven of the reports did not provide production rates and therefore could not be used to revise the emission factors. PES was able to use only one of these reports (obtained from EPA library) to revise the controlled sulfur dioxide emission factor for a three-stage catalytic unit.

A final set of reference materials was compiled after a thorough review of the pertinent reports, documents, and information according to these criteria. The final set of reference materials is given in Chapter 4.

3.2 EMISSION DATA QUALITY RATING SYSTEM

As part of Pacific Environmental Services' analysis of the emission data, the quantity and quality of the information contained in the final set of reference documents were evaluated. The following data were always excluded from consideration:

1. Test series averages reported in units that cannot be converted to the selected reporting units;
2. Test series representing incompatible test methods (e.g., comparison of the EPA Method 5 front-half with the EPA Method 5 front- and back-half);
3. Test series of controlled emissions for which the control device is not specified;
4. Test series in which the source process is not clearly identified and described; and
5. Test series in which it is not clear whether the emissions were measured before or after the control device.

Data sets that were not excluded were assigned a quality rating. The rating system used was that specified by the OAQPS for the preparation of AP-42 sections. The data were rated as follows:

A

Multiple tests performed on the same source using sound methodology and reported in enough detail for adequate validation. These tests do not necessarily conform to the methodology specified in either the inhalable particulate (IP) protocol documents or the EPA reference test methods, although these documents and methods were certainly used as a guide for the methodology actually used.

B

Tests that were performed by a generally sound methodology but lack enough detail for adequate validation.

C

Tests that were based on an untested or new methodology or that lacked a significant amount of background data.

D

Tests that were based on a generally unacceptable method but may provide an order-of-magnitude value for the source.

The following criteria were used to evaluate source test reports for sound methodology and adequate detail:

1. Source operation. The manner in which the source was operated is well documented in the report. The source was operating within typical parameters during the test.
2. Sampling procedures. The sampling procedures conformed to a generally acceptable methodology. If actual procedures deviated from accepted methods, the deviations are well documented. When this occurred, an evaluation was made of the extent to which such alternative procedures could influence the test results.
3. Sampling and process data. Adequate sampling and process data are documented in the report. Many variations can occur unnoticed and without warning during testing. Such variations can induce wide deviations in sampling results. If a large spread between test results cannot be explained by information contained in the test report, the data are suspect and were given a lower rating.
4. Analysis and calculations. The test reports contain original raw data sheets. The nomenclature and equations used were compared to those (if any) specified by the EPA to establish equivalency. The depth of review of the calculations was dictated by the reviewer's confidence in the ability and conscientiousness of the tester, which in turn was based on factors such as consistency of results and completeness of other areas of the test report.

3.3 EMISSION FACTOR QUALITY RATING SYSTEM

The quality of the emission factors developed from analysis of the test data was rated utilizing the following general criteria:

A (Excellent)

Developed only from A-rated test data taken from many randomly chosen facilities in the industry population. The source category is specific enough so that variability within the source category population may be minimized.

B (Above average)

Developed only from A-rated test data from a reasonable number of facilities. Although no specific bias is evident, it is not clear if the facilities tested represent a random sample of the industries. As in the A-rating, the source category is specific enough so that variability within the source category population may be minimized.

C (Average)

Developed only from A- and B-rated test data from a reasonable number of facilities. Although no specific bias is evident, it is not clear if the facilities tested represent a random sample of the industry. As in the A-rating, the source category is specific enough so that variability within the source category population may be minimized.

D (Below average)

The emission factor was developed only from A- and B-rated test data from a small number of facilities, and there is reason to suspect that these facilities do not represent a random sample of the industry. There also may be evidence of variability within the source category population. Limitations on the use of the emission factor are noted in the emission factor table.

E (Poor)

The emission factor was developed from C- and D-rated test data, and there is reason to suspect that the facilities tested do not represent a random sample of the industry. There also may be evidence of variability within the source category population. Limitations on the use of these factors are always noted.

The use of these criteria is somewhat subjective and depends to an extent on the individual reviewer. Details of the rating of each candidate emission factor are provided in Chapter 4 of this report.

3.4 REFERENCES FOR CHAPTER 3

1. Technical Procedures for Developing AP-42 Emission Factors and Preparing AP-42 Sections. U.S. Environmental Protection Agency, Emissions Inventory Branch, Office of Air Quality Planning and Standards, Research Triangle Park, NC, 27711, April, 1992.
[Note: this document is currently being revised at the time of this printing.]
2. Compilation of Air Pollutant Emission Factors, Volume I: Stationary Sources, Supplement A, Appendix C.2, "Generalized Particle Size Distributions." U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC, October 1986.

4.0 POLLUTANT EMISSION FACTOR DEVELOPMENT

4.1 REVIEW OF SPECIFIC DATA SETS

The uncontrolled and controlled sulfur dioxide (SO₂) emission factors in the previous version of AP-42 Section 5.18 (February 1980) were derived solely from a material balance based on the sulfur recovery efficiencies of the units. An equation used was derived from the mass balance assuming that the percent sulfur lost was equal to 100 minus the percent sulfur recovered, and that all of the percent sulfur loss is emitted as SO₂. The resulting equation is as follows:

$$_2 \text{ emissions (lb/ton)} = \frac{(100 - \% \text{recovery})}{\% \text{recovery}} \times 4000$$

Typical sulfur recovery efficiencies for uncontrolled Claus plant with two, three and four catalytic stages in the 1980 version were taken from an article in *Pollution Engineering Journal*, January/February 1972 edition (Reference #15). However, after reviewing this article, PES determined that the sulfur recovery efficiencies for an uncontrolled Claus plant with three and four catalytic stages were incorrectly reported in the previous version of Section 5.18 (February 1980). The 1972 article reports the sulfur recovery efficiencies for uncontrolled Claus plant with three and four catalytic stages as 95 to 96 percent and 96 to 97 percent, respectively. The previous version of Section 5.18 incorrectly reported the recovery efficiencies for three and four catalytic stages as 95 to 97.5 percent and 96 to 99 percent, respectively.

For this revision, PES used the average sulfur recovery efficiency for each number of catalytic stages to determine sulfur dioxide emission factors for both uncontrolled and controlled Claus plant. Therefore, the emission factors were not presented as ranges as in the previous revision, but as single values.

PES has received several new test reports from industry contacts. All of the tests received employ either a SCOT-incinerator or an incinerator alone. Reference 10 used two catalytic conversion stages and References 9, 11 and 14 used three catalytic conversion stages. References 7, 8, 12, and 13 did not specify the number of catalytic conversion stages.

Reference #7: "Compliance Test Report: Hill Petroleum Company, Texas City, Texas," METCO Environmental, Dallas, TX, December 1989.

This test was performed by METCO Environmental in accordance with EPA Reference Method 6 to determine sulfur dioxide emissions. Before being emitted, the tailgas was directed through the SCOT unit and an incinerator. The sampling location on the exhaust stack of the sulfur recovery unit was approximately 64 feet above the ground. The test did not report the number of catalytic stages used during the test, but did contain all necessary documentation and was thus rated "A."

Three runs were performed with production rates of 1.33, 1.54, and 1.31 tons per hour (TPH) and sulfur dioxide emissions of 2.50, 1.60, and 4.60 pounds per hour (PPH), respectively. The average emission factor was computed as follows:

$$[(2.50/1.33) + (1.60/1.54) + (4.60/1.31)]/3 = 2.14 \text{ pounds per ton.}$$

Reference #8: "Compliance Test Report: Amoco Oil Company, Texas City, Texas," Southwestern Laboratories, Inc., Houston, TX, May 1989.

This sampling test was performed in accordance with EPA Reference Method 6 to determine sulfur dioxide emissions from the sulfur recovery unit stack. Three 30-minute runs were performed. The tailgas was sent to a SCOT unit before it was incinerated. The test report does not indicate the number of catalytic conversion stages used during the test. However, it contains all necessary documentation and was thus rated "A." The production rate for all three runs was 18.95 tons per hour (TPH) and the calculated sulfur dioxide emissions were 19.83, 29.11, and 29.93 pounds per hour (PPH). The average emission factor is thus

$$[(19.83+29.11+29.93)/3] \text{ (PPH)} / 18.95 \text{ (TPH)} = 1.39 \text{ pounds per ton.}$$

Reference #9: "Compliance Test Report: Phillips Petroleum Company, Chatom, Alabama," Environmental Science & Engineering, Inc., Gainesville, FL, July 1991.

This test was performed in accordance with EPA Reference Method 6C. Three 30-minute test runs were conducted at the tailgas incinerator stack to determine the total sulfur emission as sulfur dioxide. Three sequential catalytic conversion stages and condensers were used to produce and collect elemental sulfur with 96 percent sulfur recovery efficiency. The average stack gas velocity, molecular weight, and moisture were determined simultaneously during each 30-minute test run. This test report was rated "A." The production rates for runs 1, 2, and 3 were 3.18, 3.04 and 3.21 tons per hour (TPH) and the sulfur dioxide emission rates were 516.8, 558.1, and 509.6 pounds per hour (PPH), respectively. The average emission factor is thus

$$[(516.8/3.18) + (558.1/3.04) + (509.6/3.21)]/3 = 168.3 \text{ pounds per ton.}$$

Reference #10: "Compliance Test Report: Collett Ventures Company, Chatom, Alabama," Environmental Science & Engineering, Inc., Gainesville, FL, May 1991.

Three 4-hour test runs were conducted at the tailgas incinerator stack to determine the total sulfur emissions. The test was performed in accordance with EPA Reference Method 6C. Method 6C was modified to measure total sulfur emissions by placing a thermal oxidizer unit prior to the analyzer. The test results yielded an average sulfur recovery of 98.57 percent. The plant employed two catalytic conversion stages to produce and collect elemental sulfur. This test report was rated "A." The throughput for runs 1, 2, and 3 were 3.74, 3.89, and 3.81 tons per hour (TPH) and sulfur dioxide emission rates were 183.30, 262.86, and 210.38 pounds per hour (PPH), respectively. The average emission factor is thus

$$[(183.30/3.74) + (262.86/3.89) + (210.38/3.81)]/3 = 57.27 \text{ pounds per ton.}$$

Reference #11: "Compliance Test report: Mobil Exploration and Producing Southeast, Inc., Coden, Alabama," Cubix Corporation, Austin, TX, September 1990.

Three 4-hour tests were performed in accordance with EPA Reference Method 6 to determine sulfur dioxide emissions. Before being emitted, the tailgas was forced through a 3-stage Claus/SCOT sulfur recovery unit. Tailgas from the SCOT unit was then piped to the incinerator where all remaining sulfur was oxidized to sulfur dioxide. Quantities of sulfur dioxide and total reduced sulfur were measured at the sulfur recovery unit stack. The test results yielded an average sulfur recovery of 99.8 percent. This test report was rated "A." The throughput for runs 1, 2, and 3 were 7.29, 6.92, and 6.96 tons per hour (TPH) and sulfur dioxide emission rates were 64.30, 41.51, and 59.53 pounds per hour (PPH), respectively. The average emission factor is thus

$$[(64.30/7.29) + (41.51/6.92) + (59.53/6.96)]/3 = 7.79 \text{ pounds per ton.}$$

Reference #12: "Compliance Test report: Hunt Refining Company, Number One Sulfur Recovery Unit, Tuscaloosa, Alabama," TTL, Inc., Tuscaloosa, AL, February 1991.

Three 60-minute test runs were performed in accordance with EPA Reference Method 6 to determine sulfur dioxide emissions. The test was conducted at the tailgas incinerator stack. This test report does not indicate the number of catalytic conversion stages used during the test. The report was given a "C" rating since the sample volume was too small. The sulfur throughput for

runs 1, 2, and 3 were 1.42, 1.46 and 1.31 tons per hour (TPH) and sulfur dioxide emission rates were 214, 145, and 159 pounds per hour (PPH), respectively. The average emission factor is thus

$$[(214/1.42) + (145/1.46) + (159/1.31)]/3 = 124 \text{ pounds per ton.}$$

Reference #13: "Compliance Test report: Hunt Refining Company, Number Two Sulfur Recovery Unit, Tuscaloosa, Alabama," TTL, Inc., Tuscaloosa, AL, August 1991.

Three 60-minute test runs were performed in accordance with EPA Reference Method 6 to determine sulfur dioxide emissions. The test was conducted at the tailgas incinerator stack. This test report does not indicate the number of catalytic conversion stages used during the test. The report was given a "C" rating since the sample volume was too small. The sulfur throughputs for runs 1, 2, and 3 were 1.35, 0.82, and 1.12 tons per hour (TPH) and sulfur dioxide emission rates were 0.87, 0.68, and 0.84 pounds per hour (PPH), respectively. The average emission factor is thus

$$[(0.87/1.35) + (0.68/0.82) + (0.84/1.12)]/3 = 0.74 \text{ pounds per ton.}$$

Reference #14: "Emission Test report: Getty Oil Company, New Hope, TX, EMB Report No. 81-OSP-9, July 1981.

Five test runs were conducted by Radian Corporation at a Claus plant with three catalytic stages. The acid gases remaining in the Claus plant tail gas were routed to an incinerator to convert the H_2S to SO_2 prior to emission to the atmosphere. The test report was given an "A" rating. EPA Method 6 was used to determine total sulfur dioxide emissions. The sulfur dioxide emission rates were: 1,318, 1,565, 1,181, 1,116, and 1,141 pounds per hour and liquid sulfur production rates were 144.7, 146.2, 123.2, 113.0, and 113.0 long tons per hour, respectively. Sulfur recovery efficiencies were 95.3, 94.6, 95.1, 95.0, and 94.9 percent. Assuming the plant operated 24 hours per day, the production rates were then 6.75, 6.82, 5.75, 5.27, and 5.27 tons per hour. The average controlled sulfur dioxide emission factor is thus

$$[(1,318/6.75 + 1,565/6.82 + 1,181/5.75 + 1,116/5.27 + 1,141/5.27)/3] = 211.68 \text{ lb/ton.}$$

This work was funded and administered by the U.S. EPA Emission Measurement Branch.

4.2 CRITERIA POLLUTANT EMISSIONS DATA

No data on emissions of volatile organic compounds, lead, nitrogen oxides, carbon monoxide, or particulate matter were found nor expected for the sulfur recovery process. Data on emissions of the remaining criteria pollutant, sulfur dioxide, is discussed below.

Sulfur dioxide.

Sulfur dioxide is the only criteria pollutant emitted from the sulfur recovery process. The previous AP-42 Section 5.18 (February, 1980) contained emission factors for uncontrolled recovery processes with two, three, and four catalytic conversion stages and one general emission factor for controlled processes without specifying the number of catalytic stages. These emission factors were not obtained from source tests, but were calculated based on the percent sulfur recovered. The equation used to calculate these emission factors was derived from material balance based on sulfur recovery efficiency. Sulfur recovery efficiencies for uncontrolled Claus plant with two, three and four catalytic stages were adopted from an article in *Pollution Engineering Journal*, January/February 1972 edition (Reference #15). PES found sulfur recovery efficiencies for uncontrolled Claus plant with three and four catalytic stages in the 1980 version were incorrectly cited. Sulfur recovery efficiencies for uncontrolled Claus plant with three and four catalytic stages were 95 to 96 percent and 96 to 97 percent, respectively. The errors had been corrected in the updated version.

Due to lack of test data, uncontrolled emission factors are unchanged in this revision. However, they were presented as single values instead of as ranges. The emission factors were calculated by taking the average of the percentage of sulfur recovered. Moreover, they were downgraded to an "E" rating since the emission factors were estimated and not derived from source tests.

PES obtained eight new source tests on controlled sulfur recovery processes. Only four test reports were used to revise the controlled emission factor for sulfur dioxide. These four tests were performed with two and three catalytic conversion stages; the others were unknown and therefore could not be used in the revision of the emission factors. Since the tests were performed with different catalytic conversion stages and at various sulfur recovery efficiencies, the results could not be averaged to give one controlled emission factor. Thus, PES has added controlled emission factors for processes with two (Reference 10) and three catalytic conversion stages

(References 9, 11 and 14) in this revision of Section 5.18. The controlled emission factors were also presented as single values instead of as ranges.

The controlled emission factors shown in Table 4.2-1 are presented for two controls: incineration and Shell Claus Offgas Treatment process (SCOT). Incineration oxidizes sulfur compounds, such as hydrogen sulfide (H_2S), carbon disulfide (CS_2), and carbonyl sulfide (COS) to form sulfur dioxide (SO_2). Incineration does not reduce the total amount of sulfur compound emissions; however, this technique does convert H_2S into less toxic SO_2 . In the SCOT process, the Claus plant tailgas is heated to 300°C (570°F) and reacted with hydrogen over a cobalt molybdenum catalyst. All sulfur compounds in the Claus reactors including sulfur vapor (S), CS_2 , and COS are reduced to H_2S . The hot gas from these exothermic reactions is cooled. The H_2S is then selectively absorbed in an amine solution. The amine is stripped, and the resultant H_2S is recycled to the front of the unit.

The controlled sulfur dioxide emission factor with two catalytic conversion stages presented in Table 4.2-1 was derived from a source test performed at the Collet Ventures facility (Reference 10). The sulfur recovery efficiency presented in the AP-42 table was calculated from the average of the percent of sulfur recovered. The emission factor is also presented as a single value. The emission factor was calculated by averaging the emission factors for the three runs:

$$[(49.01+67.57+55.22)/3] = 57.27 \text{ lb/ton.}$$

Three source test reports (References 9, 11 and 14) were used to develop the controlled sulfur dioxide emission factors with three catalytic conversion stages. Sulfur recovery efficiency presented in AP-42 table was calculated from the average of the percent of sulfur recovered from the three source tests. The emission factor however, was averaged together and presented as a single value. The average efficiency of the tests performed at Phillips Petroleum (Reference 9):

$$[.959 + .954 + .960]/3 = .958$$

The average efficiency of the tests performed at Mobil Exploration (Reference 11):

$$[.998 + .999 + .998]/3 = .998.$$

and the average efficiency of the test performed at Getty Oil (Reference 14):

$$[.953 + .946 + .951 + .950 + .949] = .950$$

Sulfur recovery efficiency for a Claus plant with three catalytic stages was derived by averaging the efficiencies of the three tests performed at Phillips Petroleum, Mobil Exploration and Getty Oil (References 9, 11 and 14):

$$[.958 + .998 + .950]/3 = .969$$

SO₂ emission factor was calculated by averaging sulfur dioxide emission factors from the three test reports:

$$[(168.29 + 7.79 + 211.68)/3 = 129.25 \text{ lb/ton.}]$$

Controlled emission factors for sulfur dioxide in the updated AP-42 document were developed from A-rated test data (References 9, 10, 11 and 14) and no specific bias is evident. However, the four facilities tested may not represent a random sample of the industries. Therefore, a "B" rating was assigned to both controlled emission factors with two and three catalytic conversion stages.

The test results show that sulfur dioxide emission factor for controlled Claus plant with three catalytic stages is higher than for Claus plant with two catalytic stages. This is probably caused by different plant design or operation.

Although References 7, 8, 12, and 13 are presented in Table 4.2-1 as A-rated source tests, they were not used to develop the emission factors because they did not indicate the number of catalytic conversion stages used during the tests. The data from these references are used only as background information.

TABLE 4.2-1 (METRIC UNITS)
SULFUR DIOXIDE

Control Equipment	Test Rating	Test Method	Run #	Production Rate ^a	Emission Rate ^b	Emission Factor ^c
Reference 7. Exhaust stack						
SCOT-absorber and incinerator	A	6C	1	1.21	1.13	0.93
			2	1.41	0.73	0.52
			3	1.19	2.09	1.76
			Average	1.27	1.32	1.07
Reference 8. Exhaust stack						
SCOT-absorber and incinerator	A	6C	1	17.2	8.99	0.52
			2	17.2	13.20	0.77
			3	17.2	13.58	0.79
			Average	17.2	11.92	0.69
Reference 9. Exhaust stack (three catalytic conversion stages)						
Incinerator (Recovery efficiency: 95.8%)	A	6C	1	2.88	234.42	81.40
			2	2.76	253.15	91.72
			3	2.91	231.15	79.43
			Average	2.85	239.57	84.18
Reference 10. Exhaust stack (two catalytic conversion stages)						
Incinerator (Recovery efficiency: 98.3 - 98.8%)	A	6C	1	3.40	83.14	24.45
			2	3.53	119.23	33.78
			3	3.45	95.43	27.66
			Average	3.46	99.27	28.63

^a Units in Mg/hr.

^b Units in kg/hr.

^c Units in kg/Mg of sulfur produced.

TABLE 4.2-1 (METRIC UNITS)
SULFUR DIOXIDE (continued)

Control Equipment	Test Rating	Test Method	Run #	Production Rate ^a	Emission Rate ^b	Emission Factor ^c
Reference 11. Exhaust stack (three catalytic conversion stages)						
SCOT-absorber and incinerator (Recovery efficiency: 99.8%)	A	6C	1	6.61	29.17	4.41
			2	6.28	18.83	3.00
			3	6.31	27.00	4.28
			Average	6.40	25.00	3.90
Reference 12. Exhaust stack						
Incinerator	A	6C	1	1.29	97.07	75.25
			2	1.32	65.77	49.83
			3	1.19	72.12	60.61
			Average	1.27	78.32	61.90
Reference 13. Exhaust stack						
Incinerator	C	6C	1	1.23	0.40	0.33
			2	0.74	0.31	0.42
			3	1.02	0.38	0.37
			Average	1.00	0.36	0.37
Reference 14 Exhaust stack (three catalytic conversion stages)						
Incinerator (Recovery efficiency: 95%)	C	6C	1	6.12	597.84	97.69
			2	6.19	709.88	114.68
			3	5.22	535.70	102.62
			4	4.78	506.21	105.90
			5	4.78	517.55	108.27
			Average	5.42	573.44	105.80

^a Units in Mg/hr.

^b Units in kg/hr.

^c Units in kg/Mg of sulfur produced.

**TABLE 4.2-1 (ENGLISH UNITS)
SULFUR DIOXIDE**

Control Equipment	Test Rating	Test Method	Run #	Production Rate ^a	Emission Rate ^b	Emission Factor ^c
Reference 7. Exhaust stack						
SCOT-absorber and incinerator	A	6C	1	1.33	2.50	1.88
			2	1.54	1.60	1.04
			3	1.31	4.60	3.51
			Average	1.39	2.90	2.14
Reference 8. Exhaust stack						
SCOT-absorber and incinerator	A	6C	1	18.95	19.83	1.05
			2	18.95	29.11	1.54
			3	18.95	29.93	1.58
			Average	18.95	26.29	1.39
Reference 9. Exhaust stack (three catalytic conversion stages)						
Incinerator (Recovery efficiency: 95.8%)	A	6C	1	3.18	516.8	162.52
			2	3.04	558.1	183.59
			3	3.21	509.6	158.75
			Average	3.14	528.17	168.29
Reference 10. Exhaust stack (two catalytic conversion stages)						
Incinerator (Recovery efficiency: 98.3 - 98.8%)	A	6C	1	3.74	183.30	49.01
			2	3.89	262.86	67.57
			3	3.81	210.38	55.22
			Average	3.81	218.85	57.27

^a Units in ton/hr.

^b Units in lb/hr.

^c Units in lb/ton of sulfur produced.

TABLE 4.2-1 (ENGLISH UNITS)
SULFUR DIOXIDE (continued)

Control Equipment	Test Rating	Test Method	Run #	Production Rate ^a	Emission Rate ^b	Emission Factor ^c
Reference 11. Exhaust stack (three catalytic conversion stages)						
SCOT-absorber and incinerator (Recovery efficiency: 99.8%)	A	6C	1	7.29	64.30	8.82
			2	6.92	41.51	6.00
			3	6.96	59.53	8.55
			Average	7.06	55.11	7.79
Reference 12. Exhaust stack						
Incinerator	A	6C	1	1.42	214.00	150.80
			2	1.46	145.00	99.70
			3	1.31	159.00	121.50
			Average	1.40	172.67	124.00
Reference 13. Exhaust stack						
Incinerator	A	6C	1	1.35	0.87	0.65
			2	0.82	0.68	0.83
			3	1.12	0.84	0.74
			Average	1.10	0.80	0.74
Reference 14. Exhaust stack (three catalytic conversion stages)						
Incinerator (Recovery efficiency: 95%)	A	6C	1	6.75	1,318	195.26
			2	6.82	1,565	229.47
			3	5.75	1,181	205.39
			4	5.27	1,116	211.76
			5	5.27	1,141	216.51
			Average	5.97	1264.2	211.68

^a Units in ton/hr.

^b Units in lb/hr.

^c Units in lb/ton of sulfur produced.

4.3 NONCRITERIA POLLUTANT EMISSIONS DATA

Hazardous Air Pollutants.

Hazardous Air Pollutants (HAPs) are defined in the 1990 Clean Air Act Amendments. HAPs such as hydrogen sulfide (H₂S), carbonyl sulfide (COS) and carbon disulfide (CS₂) are present in Claus furnace. COS and CS₂ are formed as a result of side reactions of carbon dioxide (CO₂) and the unreacted H₂S. No information or source test data are available to quantify these emissions.

Global Warming Gases.

Pollutants such as methane, carbon dioxide, and nitrous oxide have been found to contribute to overall global warming. Small amounts of CO₂ are emitted from the Claus incinerator. However, no information or source data are available to quantify CO₂ emissions. No data were found regarding methane or nitrous oxide emissions from the sulfur recovery process.

Ozone Depleting Gases.

Chlorofluorocarbons have been found to contribute to depletion of the ozone layer. No data on emissions of these pollutants were found or expected for the sulfur recovery process.

4.4 DATA GAP ANALYSIS

The amount of sulfur dioxide emitted during the sulfur recovery process varies depending upon the number of catalytic conversion stages. Only four (References 9, 10, 11 and 14) out of the eight new source test reports received indicated the number of catalytic conversion stages used during the tests. These source test reports were used to revise the controlled emission factor in the existing AP-42 document. The emission factor for controlled sulfur recovery processes with two catalytic conversion stages is derived from Reference 10 and for three catalytic conversion stages is derived from References 9, 11 and 14. PES recommends that the three plants (one plant submitted two source test reports) whose reports did not indicate the number of catalytic conversion stages used during the source tests be contacted for further information. If these four test reports cannot be used to confirm the uncontrolled emission factors for sulfur recovery processes with two, three, and/or four catalytic stages, further testing may be required. Since carbonyl sulfide and carbon disulfide emissions, both hazardous air pollutants, are known

to be emitted from the Claus furnace, emission testing is recommended to quantify these emissions.

4.5 REFERENCES FOR CHAPTER 4

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5. Review of New Source Performance Standards for Petroleum Refinery Claus Sulfur Recovery Plants, EPA-450/3-83-014, U.S. Environmental Protection Agency, Research Triangle Park, NC, August 1983.
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7. "Sampling Test Report: Hill Petroleum Company, Texas City, Texas," METCO Environmental, Dallas, TX, December 1989.
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13. "Compliance Test Report: Hunt Refining Company-Number Two Sulfur Recovery Unit, Tuscaloosa, Alabama," TTL, Inc., Tuscaloosa, AL, August 1991.
14. "Emission Test Report: Getty Oil Company, New Hope, TX," EMB Report No. 81-OSP-9, July 1981.

15. D.K. Beavon, "Abating Sulfur Plant Tail Gases," pollution Engineering, January/February 1972, pp. 34-35.

TABLE 4.4-1
LIST OF CONVERSION FACTORS

Multiply:	by:	To obtain:
mg/dscm	4.37×10^{-4}	gr/dscf
m ²	10.764	ft ²
acm/min	35.31	acfm
m/s	3.281	ft/s
kg/hr	2.205	lb/hr
kPa	1.45×10^{-1}	psia
kg/Mg	2.0	lb/ton
Mg	1.1023	ton

Temperature conversion equations:

Fahrenheit to Celsius:

$$^{\circ}\text{C} = \frac{(^{\circ}\text{F} - 32)}{1.8}$$

Celsius to Fahrenheit:

$$^{\circ}\text{F} = 1.8(^{\circ}\text{C}) + 32$$

APPENDIX A.

AP-42 SECTION 5.18

[Not presented here. See instead current AP-42 Section 8.13.]